Satellite observations of isoprene from the Cross-track Infrared Sounder

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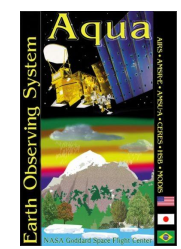
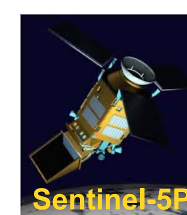
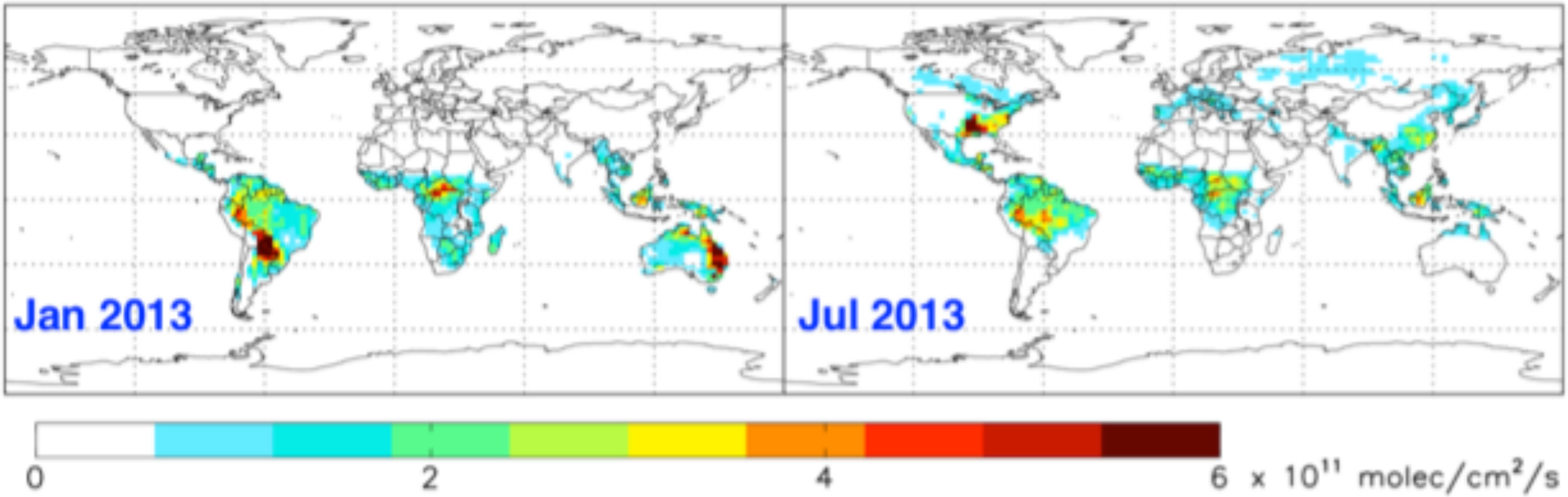




Isoprene: the dominant source of reactive carbon to the atmosphere



- major impacts of secondary organic aerosols, O_3, other oxidants
- large, highly heterogeneous emissions
- central MEGAN v2.1 bottom-up estimate: ~470 TgC/year

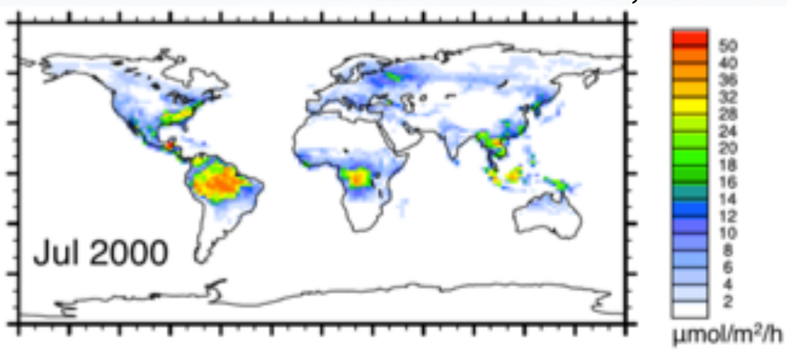
vs.

  - EDGAR total anthropogenic VOC emissions: ~160 TgC/year (*Huang et al.*, 2017)
  - global methane emissions: ~560 TgC/year (*Saunois et al.*, 2016)

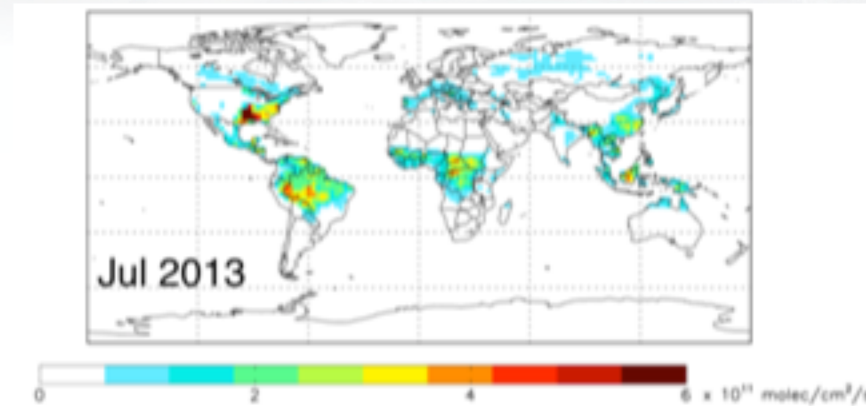
Wide disparity between bottom-up isoprene flux estimates

Central MEGANv2.1 estimate:
470 TgC/year

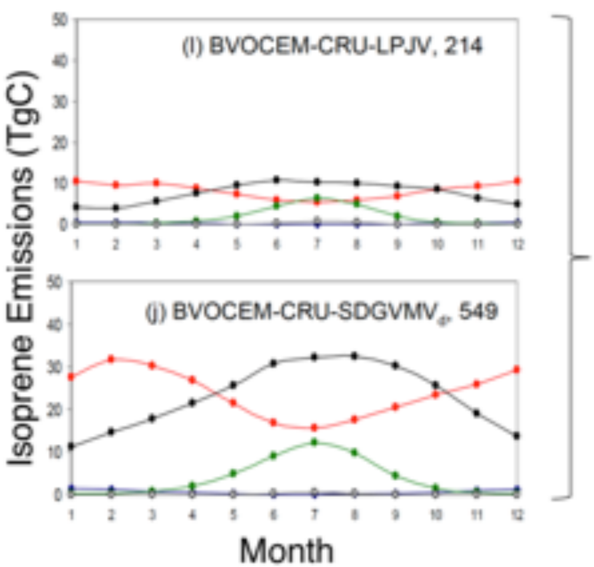
Guenther et al., 2012



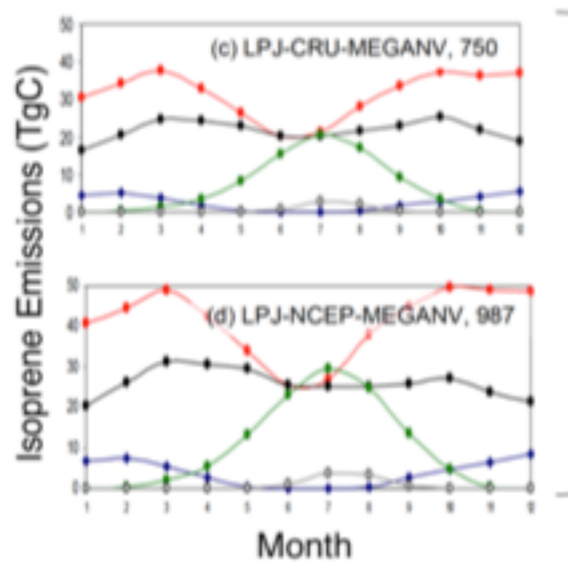
MEGANv2.1 as implemented in GEOS-Chem:
206 TgC/year



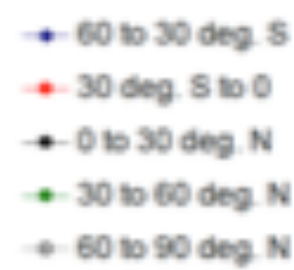
Strong sensitivity to model meteorology, land cover (vegetation type, leaf area), canopy parameterization …, besides the built-in emission algorithms



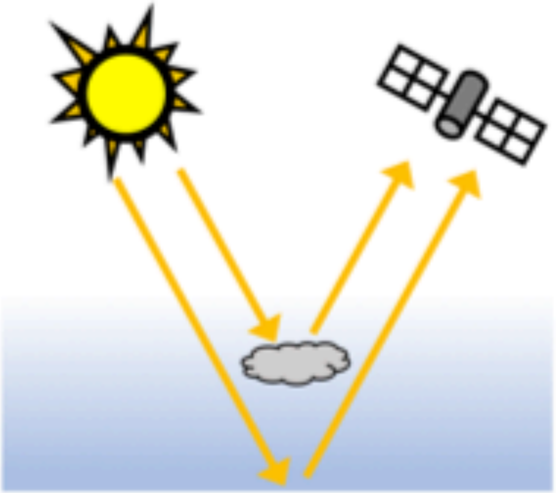
Same emission model & meteorology; different vegetation
214 vs 549 TgC/year



Same emission model & vegetation; different meteorology
750 vs 987 TgC/year

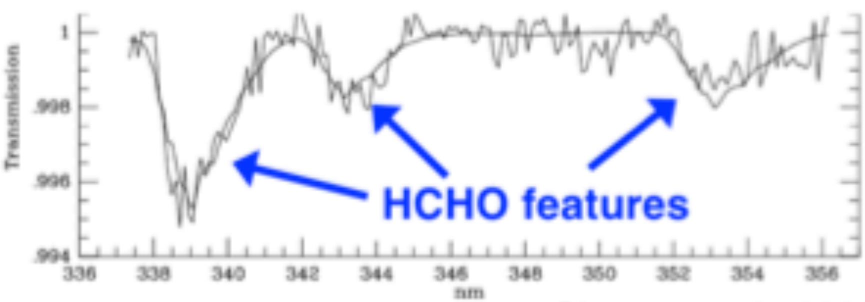


HCHO measurements from space provide top-down constraints



HCHO near-UV absorption features permit remote measurement via solar backscatter

HCHO absorption as seen by GOME-1 for a scene over the SE US

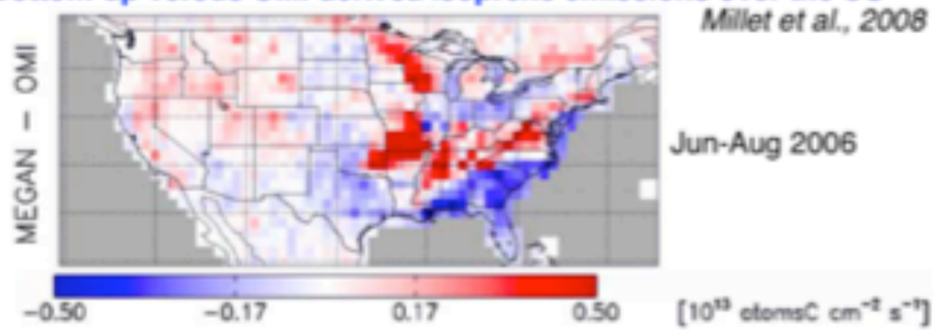


Chance et al., 2000

HCHO is produced in high-yield from isoprene oxidation, and HCHO column measurements have informed our understanding of isoprene emissions.

Bottom-up versus OMI-derived isoprene emissions over the US

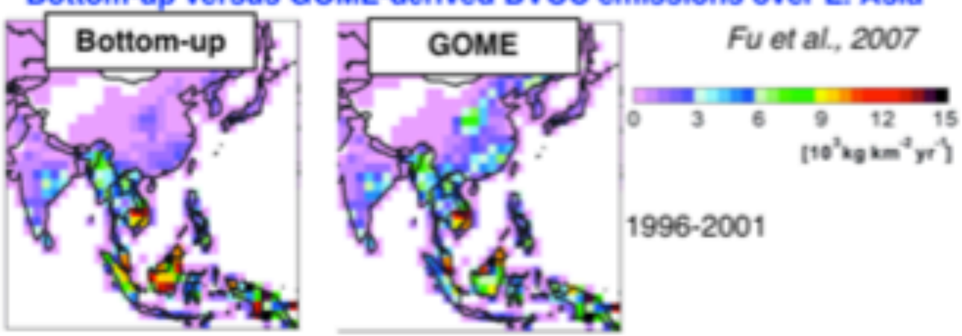
Millet et al., 2008



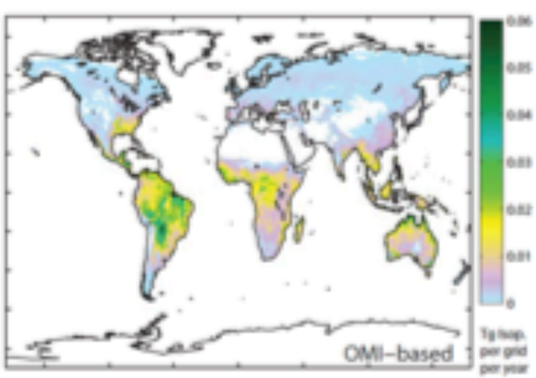
Jun-Aug 2006

Bottom-up versus GOME-derived BVOC emissions over E. Asia

Fu et al., 2007



1996-2001



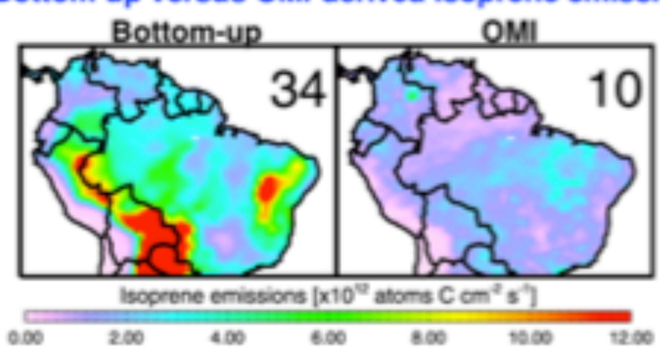
Global isoprene emissions derived from OMI

Bauwens et al., 2016

2005-2013

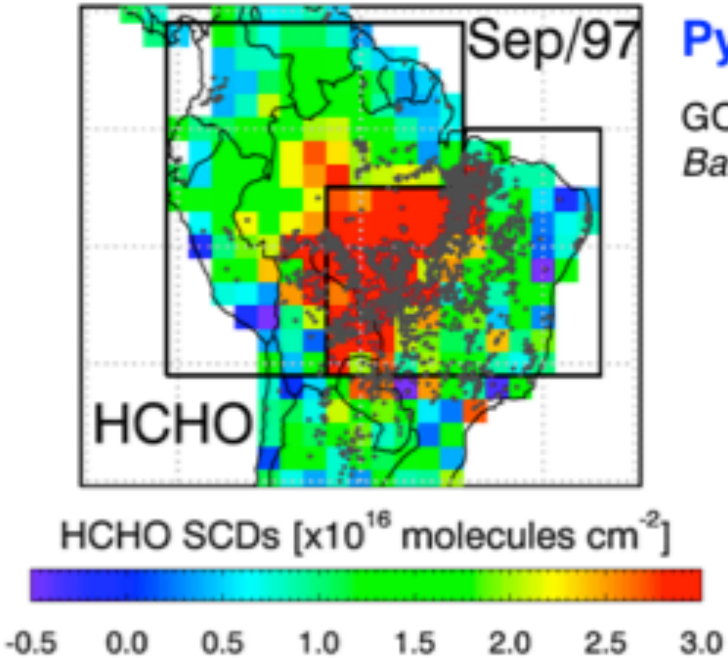
Bottom-up versus OMI-derived isoprene emissions over Amazonia

Barkley et al., 2013



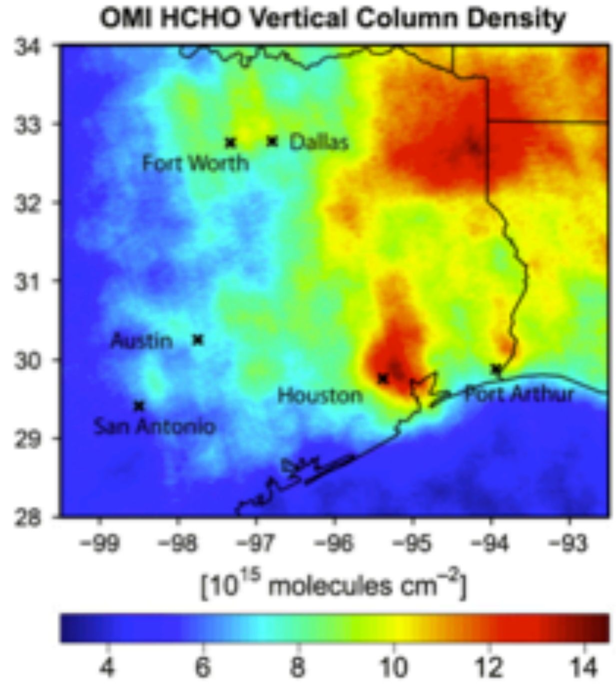
Jan. 2006

Other HCHO sources, chemical complexities challenge interpretation



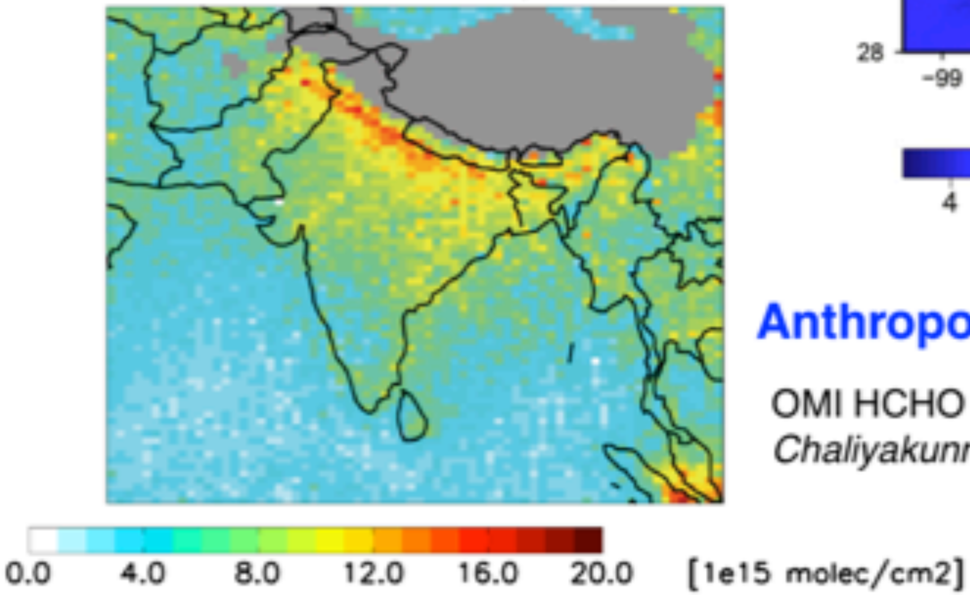
Pyrogenic VOCs

GOME HCHO columns with ATSR fire counts overplotted
Barkley et al., 2008



Anthropogenic VOCs

OMI HCHO enhancement over Houston
Zhu et al., 2014

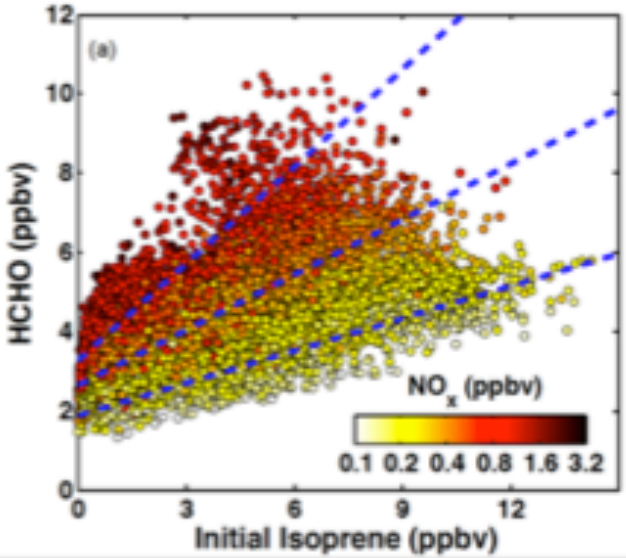


Anthropogenic, pyrogenic, and biogenic VOCs

OMI HCHO enhancement over the IGP
Chaliyakunnel et al., in revision

Other HCHO sources, chemical complexities challenge interpretation

HCHO:isoprene relationship varies with OH, NO_x



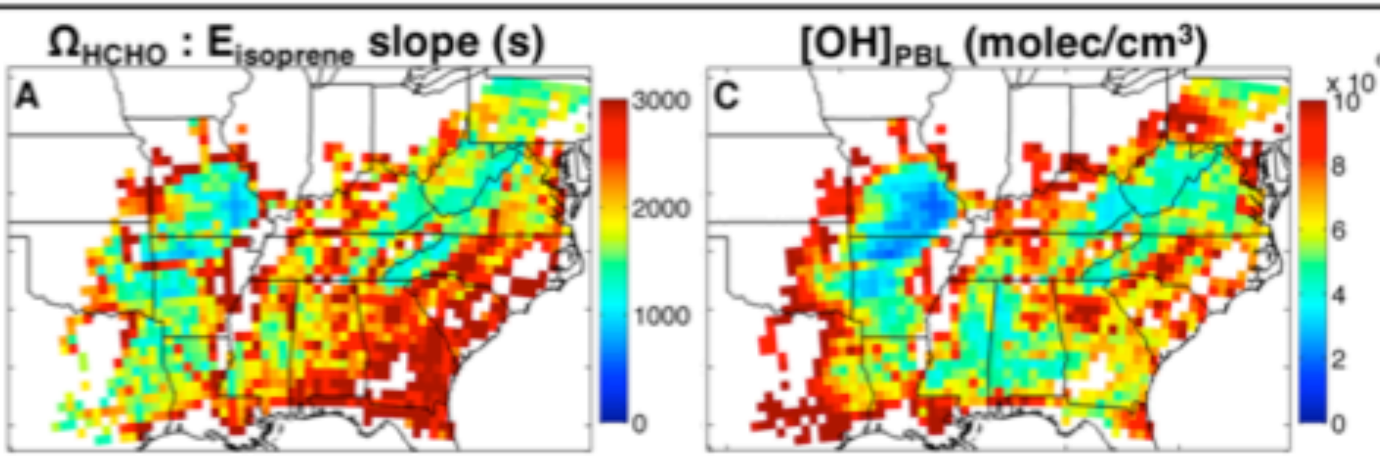
Initial isoprene versus resulting HCHO as a function of NO_x during SENEX *Wolfe et al., 2016*

Prompt HCHO yield increases 3× from low to high NO_x

$$P(HCHO) = \alpha \cdot P(RO_2)$$

Increase branching ratio (HCHO yield)

Increase radical cycling ($HO_2 \rightarrow OH$) and ∴ $P(RO_2)$

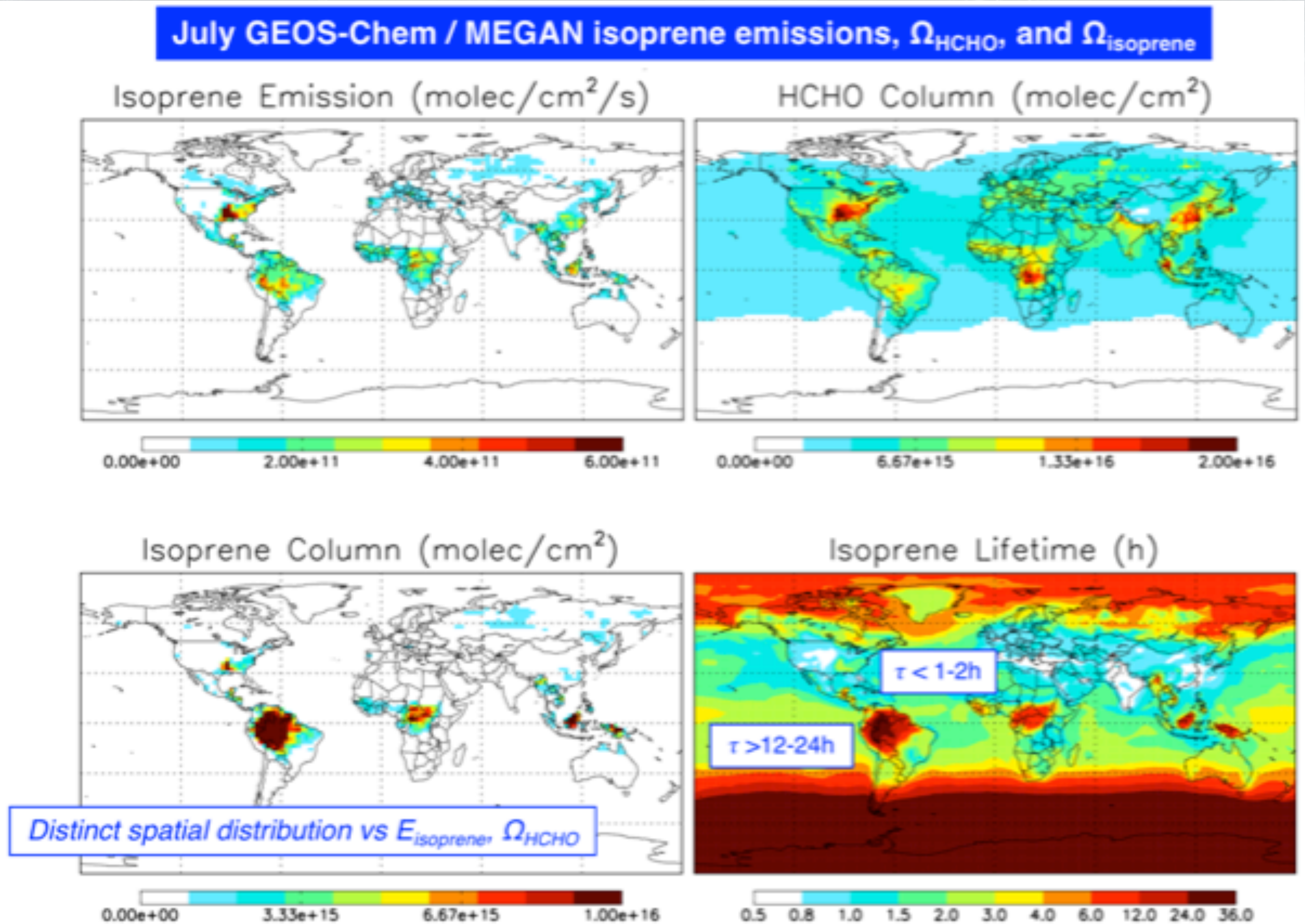


Modeled HCHO:isoprene relationship modulated by OH

Valin et al., 2016

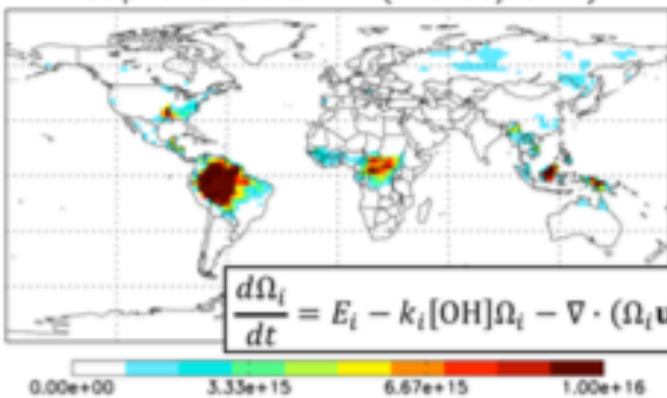
Using Ω_{HCHO} to constrain isoprene sources: we rely on CTMs to accurately capture these effects

Concurrent Ω_{HCHO} and $\Omega_{isoprene}$ measurements would help constrain isoprene emissions and its chemistry

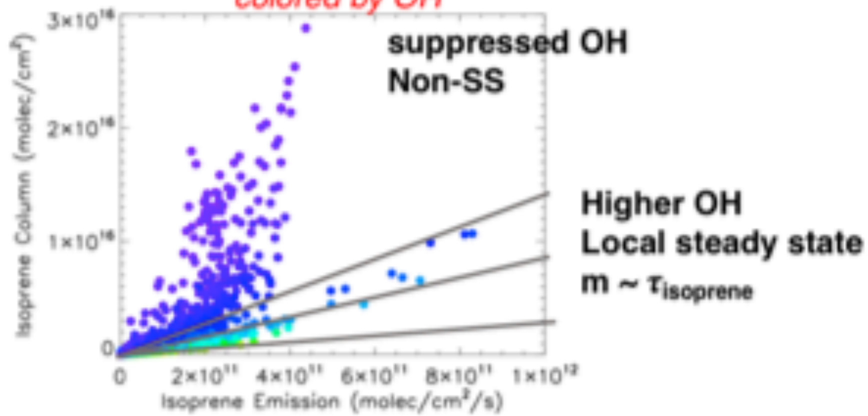


Concurrent Ω_{HCHO} and $\Omega_{isoprene}$ measurements would help constrain isoprene emissions and its chemistry

Sensitivity of $\Omega_{isoprene}$ to OH regime & chemistry



$$\frac{d\Omega_i}{dt} = E_i - k_i[\mathrm{OH}]\Omega_i - \nabla\cdot(\Omega_i \mathbf{u})$$

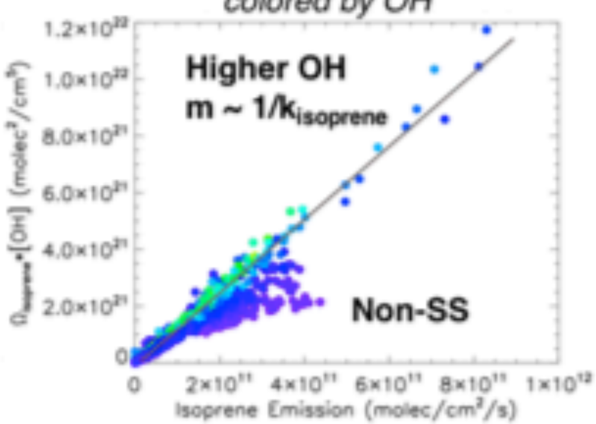


Ω_{HCHO} – more buffered to OH changes

$$\frac{d\Omega_f}{dt} = \alpha k_i[\mathrm{OH}]\Omega_i - (k_f[\mathrm{OH}] + J_f)\Omega_f - \nabla\cdot(\Omega_f \mathbf{u})$$

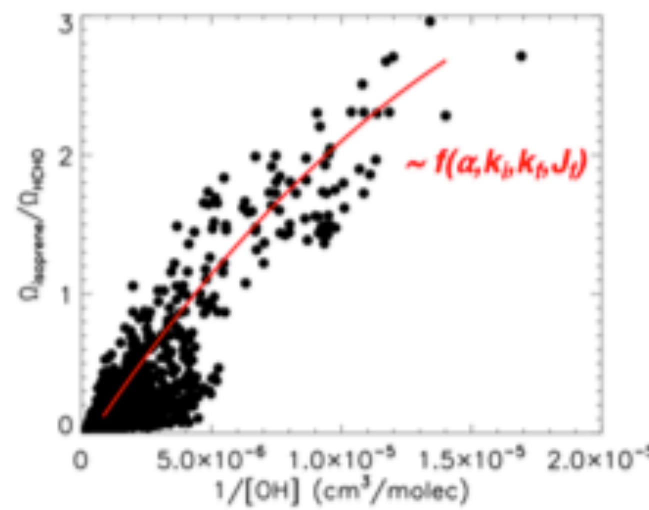
L(HCHO) buffered due to photolysis

P(HCHO) ~ [OH]Ω_i, less sensitive to OH change than is Ω_i



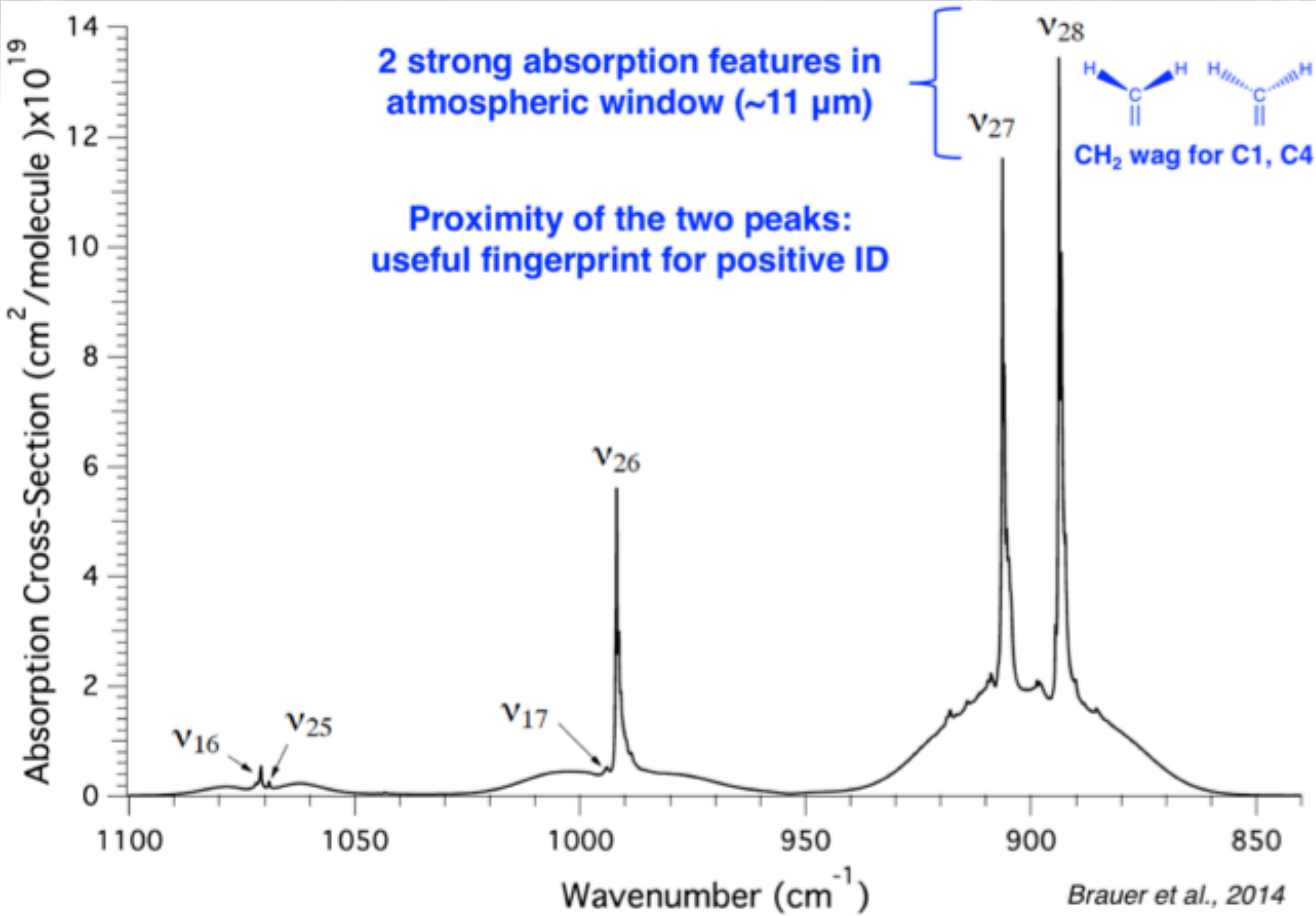
** caveat: 2x2.5° model run!*

$\Omega_{isoprene}$ / Ω_{HCHO} – scales closely with OH

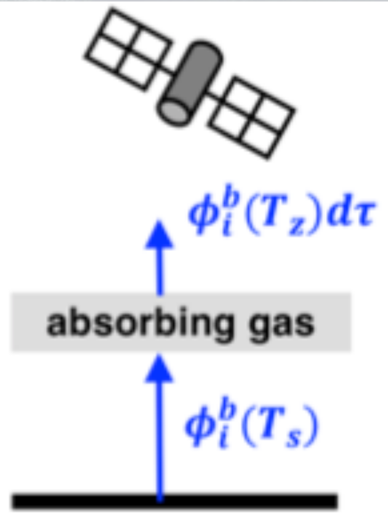


$\Omega_{isoprene}$, Ω_{HCHO}: complementary information on isoprene sources and chemistry (i.e., OH suppression)

Thermal infrared spectroscopy of isoprene



Feasibility of isoprene measurements using space TIR spectrometers



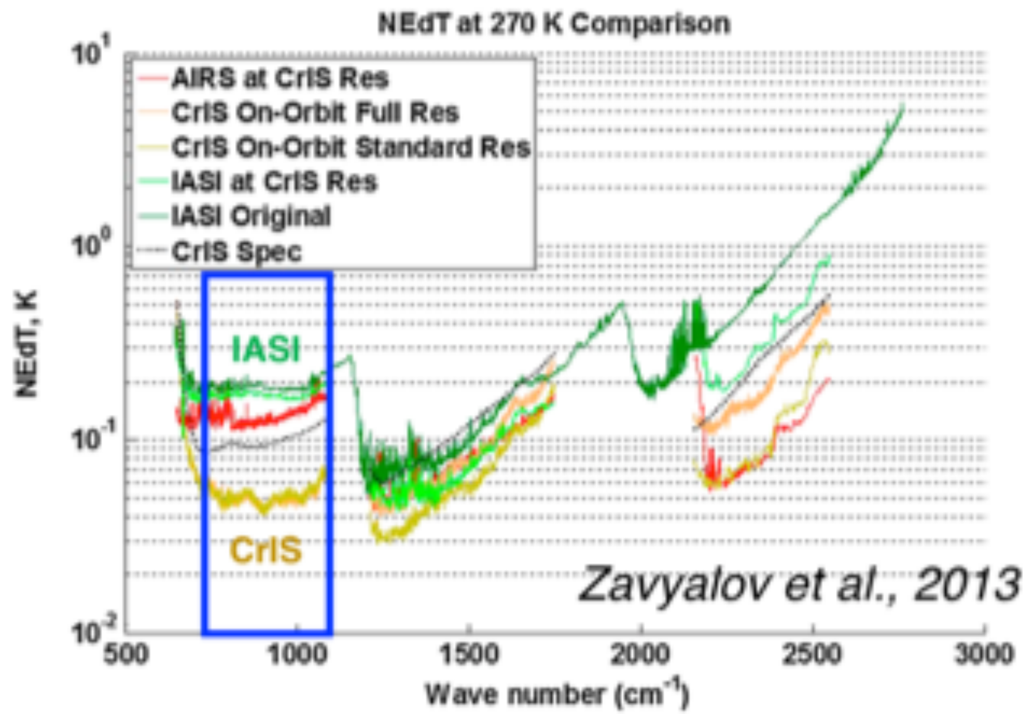
Challenges

- mainly resides in near-ground since its emission sources at surface and its short life time
- its weak spectral signature, interfered by other species (H_2O, HNO_3, NH_3, CFCs)

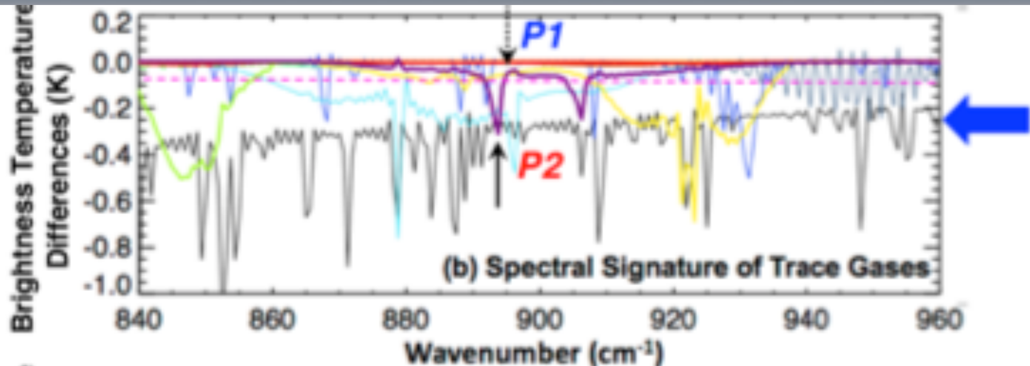
CrIS

- 1:30 pm local time overpass when isoprene, thermal contrast, vertical mixing are high
- Lower noise than the other space sensors
- Fine spatial resolution & global coverage enabling the spatial/temporal averaging to achieve the desirable signal to noise ratio for enabling isoprene retrievals

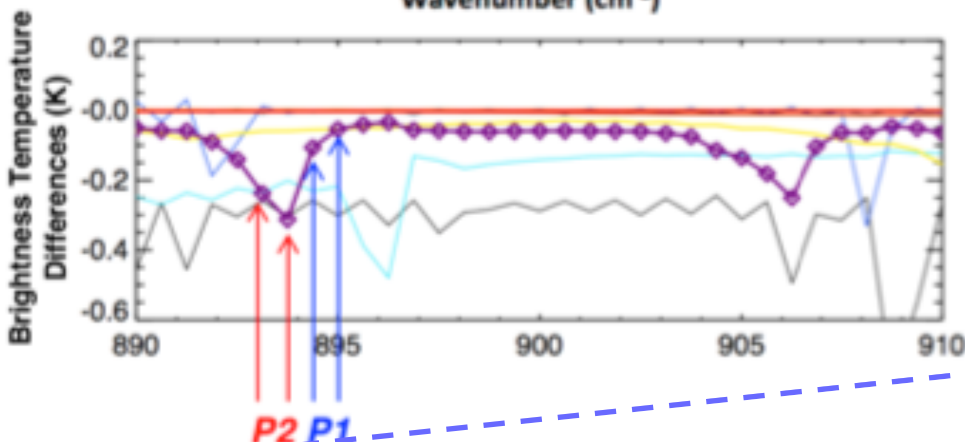
Noise characteristics of CrIS, IASI, AIRS



Atmospheric isoprene spectral signature detected by CrIS

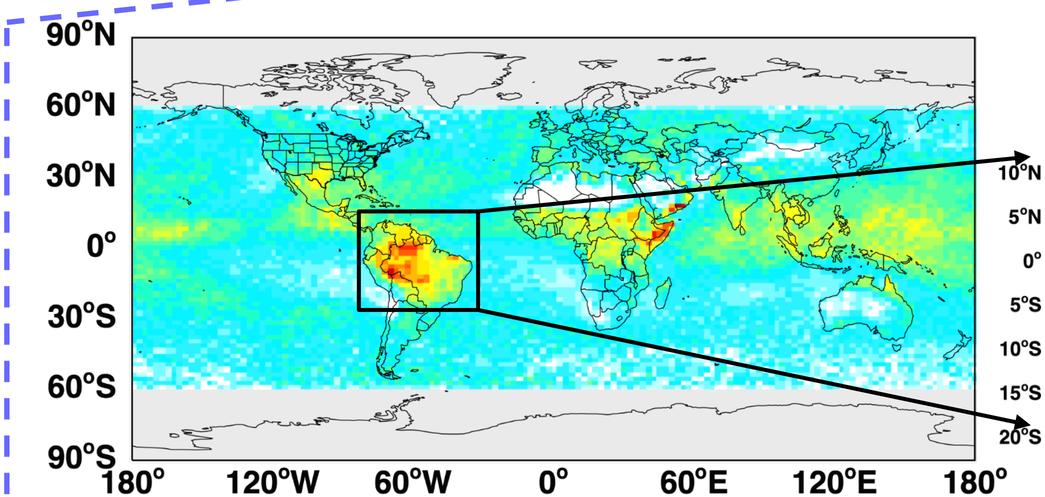


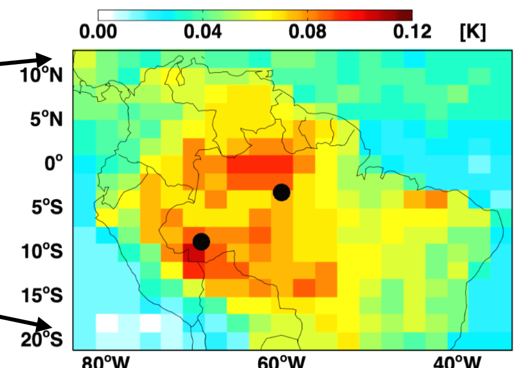
- Species within the spectral region of interest: **Isoprene, H_2O, CO_2, NH_3, HNO_3, CFC11, CFC12**
- Isoprene spectroscopic parameters from Brauer *et al.* (2014)
- Isoprene u_{28} band better separated from the interfering species

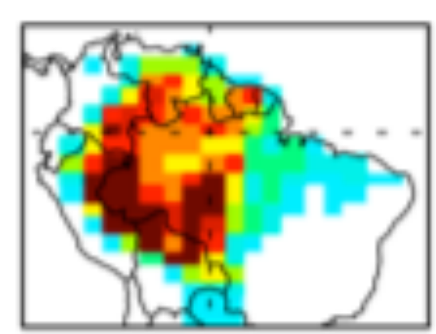


Rapid verification of the isoprene signals using brightness temperature difference (△BT) approach:

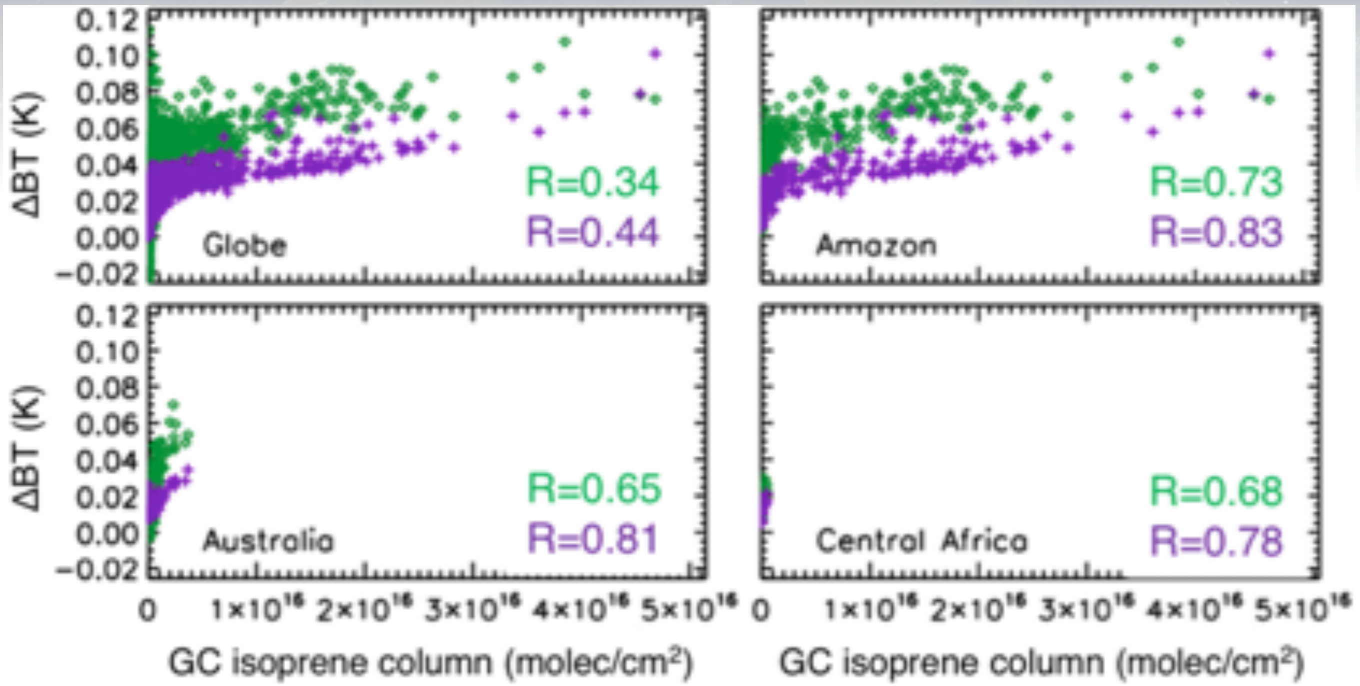
- △BT = radiance@P1 – radiance@P2







△BT distribution reveals presence of isoprene signatures



Monthly averaged signals vs. GEOS-Chem isoprene (C_5H_8) columns for September 2014

Green diamonds: △BT observed by CrIS vs. GEOS-Chem predicted isoprene

1) **△BT correlates with predicted C_5H_8 columns reveals the presence of C_5H_8 signature in CrIS measurements**
2) **Higher isoprene amount over Amazon region than other regions**

Purple plus: predicted relationship using a radiative transfer model (RTM)

1) **Observed △BT-isoprene correlation matches theoretical expectation**
2) **The △BT offset of the predicted relationship vs. CrIS measurements (off-peak - peak radiances), suggest spectral interferents not yet fully represented in the RTM simulation since the simulation does not use the instantaneous atmospheric state, surface and cloud properties. --- While would not be an issue if conducts full physical retrievals of isoprene and interferences.**

Approaches for quantifying isoprene columns from CrIS

Full physical retrievals

Developed upon the MUSES full physical retrieval algorithm (Fu et al., 2013; 2016; 2018)

Apply optimal estimation to quantify the isoprene amounts that best fits CrIS radiances co-retrieving atmospheric state, surface/cloud properties, as well as the a priori isoprene profiles

Pros: (1) detailed sensitivity and uncertainty information for each measurement

(2) take the spectral interferences of gases and surface/cloud properties into account

Cons: (1) demands of computation resources; *could be mitigated/addressed via using fast RTM (~23X reduction in computation time), target scenes selections (skipping both cloudy and nocturnal scenes; ~3X reduction)*

(2) impacts of a priori constraints; *does not show significant impacts on the retrieved columns*

Artificial neural network

Train an ANN based on simulated data, based on the approach used for IASI NH_3 [Whitburn et al., 2016]

Apply ANN to predict isoprene columns from observed CrIS $\triangle$BT and other relevant parameters

Dr. Kelley Wells (UofM) is developing an ANN for CrIS Isoprene [Details available in the coming AGU Fall 2018 & AMS 2019 conferences]

Pros: (1) fast -- negligible computation time

(2) does not use a priori constraints

Cons: (1) does not account for variable sensitivity among target scenes

(2) purely empirical approach, lacks of link between radiances and isoprene amounts

(3) the impacts of accuracy/precisions of parameters used in the prediction

Full physical retrievals using MUSES algorithm

Step 1: Retrieve temperature, surface/cloud properties, and abundances of other species

△BT from this pre-isoprene retrieval step

=

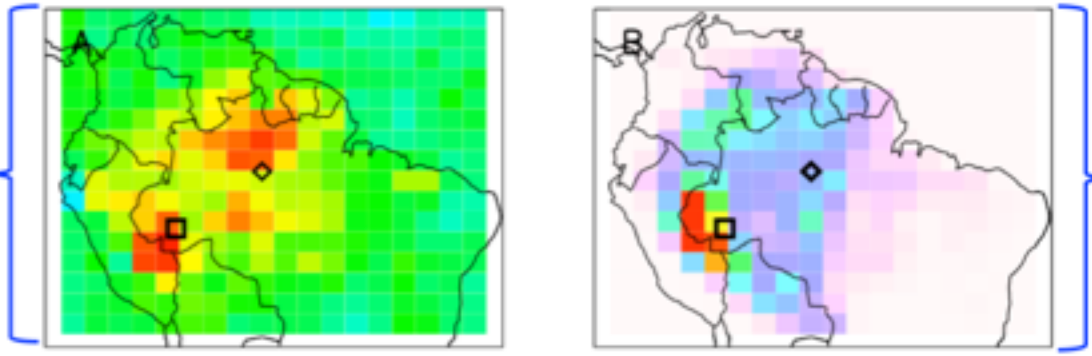
Simulated BT without isoprene – CrIS measurements

Note:

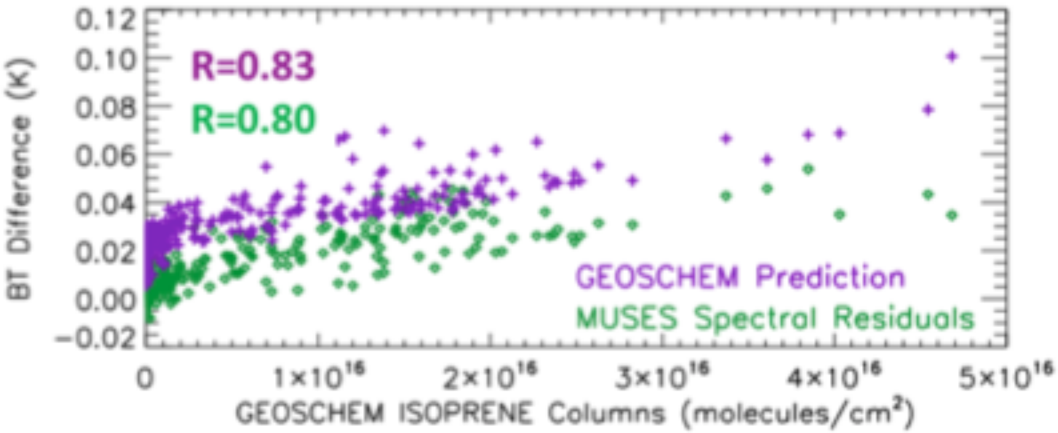
It is an approach different than the △BT estimation of *off-peak vs. peak radiances.*

It accounts for the interferences of instantaneous atmospheric state, and surface/cloud properties for each CrIS measurement.

September 2014



GEOS-Chem isoprene columns



△BT from pre-isoprene retrieval step

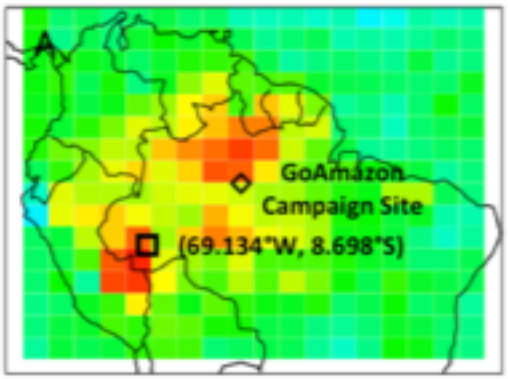
1) meets theoretical expectation
2) reports △BT ~= 0 (Y axis intercept) when
3) takes the spectral interferences of other species, land/cloud properties

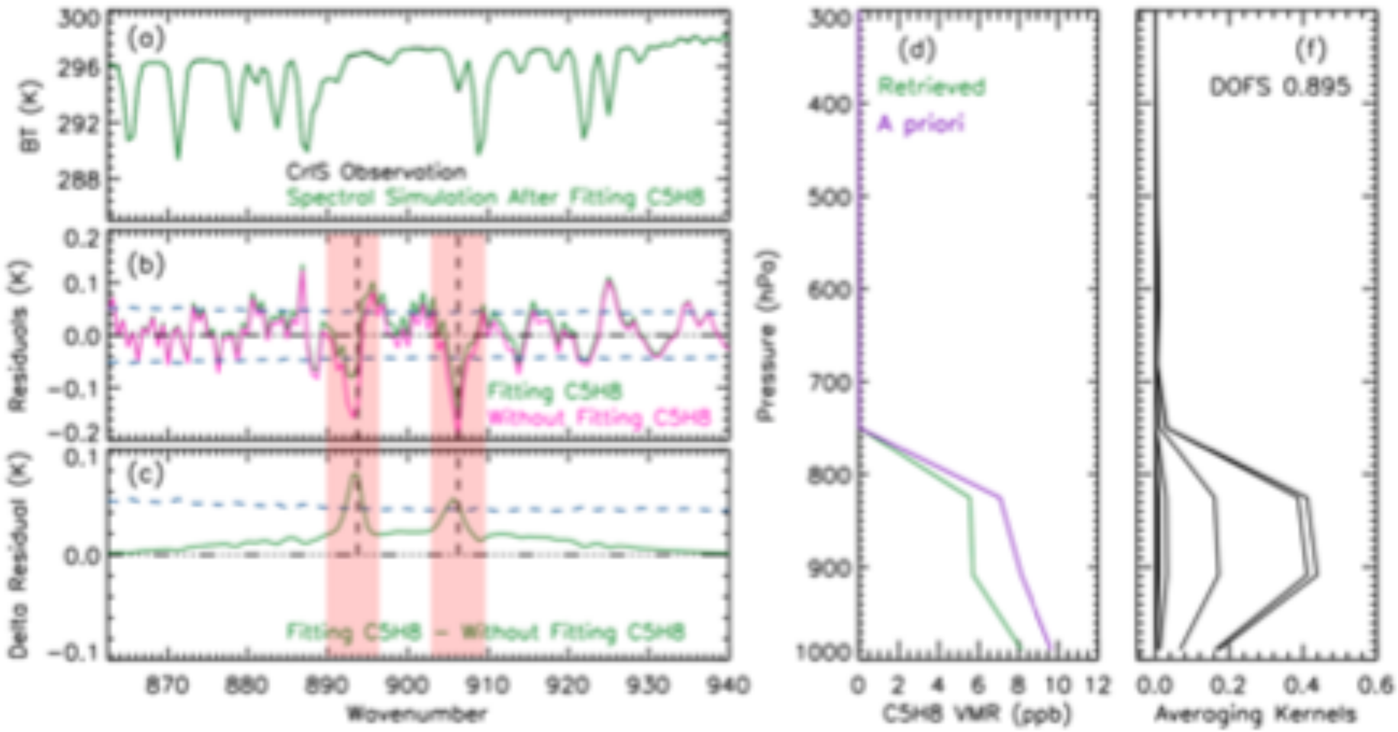
Full physical retrievals using MUSES algorithm

Step 2: Retrieve isoprene

<u>Prototype CrIS isoprene retrieval</u>

Single measurement at (69.134W, 8.698S) on September 12, 2014





Ongoing

1) conduct step 2 retrievals over Amazon for all September 2014 data, and other seasons
2) apply the optimized MUSES algorithm over the globe

Acknowledgements

Brian Drouin, Keeyoon Sung, and Timothy Crawford for providing the laboratory evaluation of the current-state of isoprene spectroscopic parameters.

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